Capped colloids as light-mills in optical traps

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Abstract. Custom-designed colloidal particles in an optical tweezers act as light-mills in a fluid. In particular, aqueous suspensions of capped colloids, in which half of the surface is covered with metal layers, are investigated. Due to their asymmetry, the capped colloids can act as rotators when exposed to intense laser fields. Particles of 4.7 μm in diameter are observed rotating around the focus of a laser beam. For low intensities, particles become trapped close to the spot of highest laser intensity. Above a threshold value of about 4 mW in total beam intensity, the particles move away from the center of the focus and start to rotate at frequencies of about 1 Hz. The balance of forces due to light pressure and hydrodynamic forces gives a constant rotation rate. The speed of the spinning particle increases linearly with laser power to above 2 Hz until the particles are ejected from the focus for intensities higher than 7 mW. Magnetic caps introduce further possibilities to tune the rotation rates.

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1. Introduction

Laser tweezers allow microscale particles to be positioned with high precision and reproducibility. The main advantage of this technique is its versatility. It can be performed in almost any transparent medium, especially in cells containing a liquid. Thus, a large number of systems (ranging from biological samples such as cells to colloidal particles) can be manipulated using this technique. A well-defined rotation of the particles can be used to further investigate particles on the micron and even nanometer scale. Therefore a large number of experiments has been performed during the past years in order to investigate the rotational motion of such particles. Typical approaches used the absorption of polarized light that transfers spin or angular momentum and refraction of the beam by custom-made objects. Rotation around the beam axis [1, 2] and perpendicular to it [3] could be studied and theoretically explained [4]. Additionally, turning the angle of polarization of an optical tweezers can also lead to rotation of birefringent particles [5]. Wherever light pressure propels the motion of particles, the rotating object needs to be asymmetric. In the system investigated here, asymmetry is introduced by coating one side of the spheres with metal caps. Visualization of Brownian motion of similar particles [6] and rotation of particles with magnetic caps in a magnetic field have already been demonstrated [7]. Accordingly, charged particles with metal caps move in an electric field [8]. More detailed studies on rotation in optical traps use biochemically joined particles as dimers [9] and pieces of glass powder [10] as rotating agents. In this work, we describe the controlled off-axis rotation of capped colloids around a Gaussian laser spot is described.

2. Sample preparation and experimental setup

The samples, named capped colloids, were prepared in the process illustrated in Figure 1. It starts with the production of a monolayer, which is similar to the method described in [11]. A drop of suspension with 4.7 µm Silica particles (SS05N from Bangs Laboratories) totally wets a thoroughly cleaned borosilicate glass cover slip. It dries in a cell at water saturation pressure, which is tilted by about 5°. Through self-assembly during evaporation of the liquid, the particles arrange in hexagonal order.
The sample is then transferred into an evaporation chamber and a coating is added. After an adhesive layer of $1-2$ nm Ti, various metals such as Ni, Au or Co-Pd multilayers are deposited on the particles [12]. The final coating, added by e-beam evaporation, consists of a SiO_x layer, which should guarantee uniform surface properties such as surface charge.
To detach the particles from the substrate, a stepper motor stage dips the cover slips into a water basin under 45° at about 10 µm/s. Surface tension peels the colloidal particles off the substrates so that they float on top of the water surface [13]. After pipetting the excess fluid from the reservoir, the particles are mixed with the residual liquid. Using this recipe, they disconnect from each other and give suspensions of variable concentration in capped colloids. Alternatively, ultrasound also removes particles from the surface [14].
Finally, the suspensions are confined between two glass plates separated by an O-ring. The lower glass slip is coated with a PMMA layer to provide a smooth surface, which prevents sticking of the particles. For the same reason, the particles are suspended in deionized water.
Such sample cells are examined in an optical tweezers setup. The colloidal spheres are imaged with an inverted video microscope onto a CCD camera, and particle positions are determined with a rate of 1 Hz and a lateral accuracy of 100 nm. From these data we obtain particle positions and the relative orientations of the caps.
A frequency doubled Nd:YVO4 laser ($\lambda = 532$ nm) generates the Gaussian shaped laser focus with a full width at half maximum of about 2.8 µm. The light is collimated

through a 20x objective placed directly above the sample cells. Rotation rates were determined with a photo diode mounted at one side of the sample cell by recording the sequence of light pulses.

3. Results

The orientation and rotational motion of capped colloids are clearly visible. This allows to characterize the system in a controlled and precise way.
Figure 2 depicts capped colloids rotating in the optical trap. The laser beam driving the motion of the particles is reflected by the inner and outer cap surfaces. The light-house effect results from this reflection at the cap. It becomes visible in the images because of the scattering of the reflected light from the substrate surface. Figure 2a and a video in the supporting material visualize this effect. In order to determine the orientation of the cap in greater detail, the intensity of the laser light in front of the camera is reduced. This reveals that the motion of the particle is eccentric. It moves on a circle with a diameter of about 1.5 µm around the focal point. Although the exact orientation of the cap is not immediately obvious, it is inferred from the images that its edge aligns vertically. So the plane separating the coated and uncoated parts lies perpendicular to the substrate. Tilting a cap away from this position lowers the rotation rate until the particle stops. This can be achieved by an external field interacting with a magnetic cap. There is no preference either for rotation in clockwise or counterclockwise directions and jumps between them occur spontaneously.
To induce the rotation of the particles, a threshold value in laser intensity needs to be overcome. Below this intensity, the capped particles can be trapped and moved, but do not rotate. As shown in Figure 3, above this value the rotation rate increases almost linearly with the laser power. Each individual particle shows a monotonous increase in rotation rate with laser power. The errorbars indicate the distribution of the frequencies for an ensemble of particles. The standard deviation in rotation rate for one specific capped colloid is approximately the symbol size. At a certain point (here $\sim$ 7.5 mW), the light pressure is high enough to expel particles from the laser focus.
In further investigations, the mechanism of the rotation was investigated in more detail. In particular, the relative positions of the laser tweezers and the particle center have been determined. At first, the position of the laser focus was determined from a picture without the particle. Subsequently, colloids and caps were located in images resulting after a threshold in brightness level had been applied. Dark areas corresponding to particles in the pictures were identified and located by edge detection. The position of the cap was found as the barycenter of the dark region. With this information, the normal vector of the caps could also be determined. It was given by the relative position of the particle center and the barycenter of the cap. For each particle the distance to the beam center was determined. Finally, the relative coordinates were converted into the coordinate system of the capped colloid. The resulting distribution of focal positions is depicted in Figures 4c, f.
An effective potential for the asymmetric particles can be given and illustrates the bistability of the system. The Boltzmann equation is used to convert the frequency of relative positions as displayed in Figures 4a, d into a potential energy landscape. Only the radial dependence of the effective potential is retained. The angle between the cap and the vector from the particle center to the position of the laser focus has been integrated out. For uncapped particles, the potential has a minimum at the origin and increases with radius as shown in [15]. For capped particles, the pivotal minimum in the radially averaged potential shifts to values around 0.5 µm as seen in Figure 4b. Here, particles are simply held in the tweezers. The potential further develops a second minimum for higher laser powers as in Figure 4e. For increasing intensity, the dip at a distance of 1.5 µm from the center of the laser focus becomes more and more pronounced. When the particles are trapped in this state, they rotate around the focus.

The orientation vectors for the caps always point against the direction of the motion. Brownian motion can lead to spontaneous changes in direction from clockwise to counterclockwise or vice versa. In the reference frame of the particles in Figure 4f, this corresponds to a position of the focus which is more to the left or more to the right of the center, respectively. The points close to the middle correspond to usual tweezing as in the case of lower intensity in Figure 4c. The ring-like distribution of the dots is due to the limited resolution of the camera, which leads to an artificial quantization of the relative positions of the particles.
Another approach to find out more about the behaviour of capped colloids in optical traps is to use magnetic caps. All capped colloids rotate irrespective of the intrinsic magnetization in the caps. Without a magnetic field, we found no difference in the interaction of the laser with non-magnetic colloids. In the absence of a magnetic field, particles having an in-plane magnetization from a 50 nm Ni cap or a perpendicular magnetization from CoPd-multilayers rotate as expected. When a magnetic cap is, however, tilted in the beam direction by an external field from two Helmholtz coils, the rotation slows with increasing magnetic field strength. Small magnetically stabilized clusters or short chains of particles can be rotated as well. Often CoPd-covered particles form ensembles of three particles so that their caps are pointing toward their center. Such clusters with a larger hydrodynamic radius rotate more slowly than individual particles. The reflectivity of the caps can be tuned by layer depth and choice of material. A minimum layer depth of about 20 nm of metal coating is needed to obtain a significantly reduced transmissivity of the capped side. Particles with thinner caps do not rotate.

4. Discussion and comparison with similar systems

From a comparison with experiments on specially designed particle dimers in [9], it is assumed that light pressure drives the system. Employing capped colloids as rotators, however, simplifies the geometry of the scattering object. For capped colloids, several forces contribute to the proposed rotation mechanism. Due to the scattering force, their transparent part is drawn into the focus of the laser tweezers. On the other hand, the momentum transfer from internal and external reflections at the coated surface pushes the cap. Both components balance at an equilibrium distance from the beam center. This leads to the second minimum in the effective potentials.
Furthermore, a torque on the particle results because of the asymmetric orientation of the cap towards the beam axis. Therefore, the angular velocity increases until it is balanced by hydrodynamic forces. Due to this asymmetric position, higher torques can be exerted compared to particles rotating about an axis through their center. To estimate the net torque from the viscous drag on the particle, simplifications in analogy to [9] yield a value of about $2.4 \cdot 10^{-18}$ Nm.
The rotation of capped colloids is robust for various choices of inner cap material (Al, Au and thin layers of C), given that Au and SiO always cover the outer surface. This supports the intuitive conclusion in [9] that the light pressure due to reflection at the outside shell drives the motion.
During rotation, irregularities in the particle motion occur. Since Brownian motion leads to a change in the direction of rotation of the particles, it might also generate fluctuations in the rotation rates. For constant laser intensity, it is expected that smaller particles will rotate faster at fixed laser intensity, as is reported for chunks of glass powder [10]. Contrary to capped colloids, however, the size and shape of such irregular particles is hard to characterize.
The manipulation of rotating capped colloids with a magnetic field shows that indeed the scattering geometry determines the rotation rate. It can be used to adjust the orientation of the particles during rotation. The more the cap is tilted, the slower does the particle rotate.

5. Conclusions and Outlook

In summary, capped colloids rotate around the focus of an optical tweezers with tunable angular velocity. Above a threshold, the particles rotate eccentrically with a rotation rate proportional to the laser intensity. Rotators can be fabricated by floating the capped particles off the substrates after evaporation of the metal layers. This prevents damage due to ultrasound and may facilitate treatments such as biofunctionalization. The rotators can be moved around and actuated by a simple optical trap. Light pressure pushes the particle in an equilibrium distance from the beam center. Effective potentials are derived from the relative positions of the trap and the particle. They visualize the transition from normal tweezing to rotation.
This design allows to exert torques for pumps, motors or drills on the micron-scale. Making use of the magnetic properties of the particles may become a further step towards micro-machines. A feedback loop for the laser power could be used to further control the rotation speed as in [10]. Additionally, a magnetic field can set the orientation of magnetic caps.

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Figure 1. Preparation of capped colloids. **a** A 30 µL drop of a 10 % solids stem suspension of 4.7 µm Silica particles dries on a clean cover slip. The particles arrange into a monolayer and diverse metals are evaporated on top. After dipping into a water reservoir the layer of particles detaches from the substrate and floats on the fluid surface. Finally, stirring breaks the particle bonds and the suspension is enclosed in transparent samples cells. **b** SEM image of two 4.7 µm Si particles on borosilicate glass object slides that have been coated with 50 nm Ni, 50 nm Au and 20 nm SiO_x layers.

Figure 2. Counterclockwise and clockwise rotation of capped colloids around a laser focus. **a** 4.7 µm Silica colloids with a 50 nm Au and 20 nm SiO_x cap rotate counterclockwise around the focus of a laser beam of 5.44 mW in an aqueous suspension. The inner cap surface reflects the light of the optical trap similar to a mirror reflecting a signal fire in a light-house. Additionally, a supplementary 1.3 MB mpeg1 is available online. **b** Filtering out most of the laser light at 4.25 mW for a clockwise rotating particle reveals that it is held eccentrically. The transparent half without cap points into the direction of motion. In both cases, the time difference between successive pictures is approximately 0.3 s.

Figure 3. Onset and frequency of rotation. Below a threshold of 4 mW of total laser intensity in the optical tweezers 50 nm Au and 20 nm SiO_x capped colloids can be trapped. Above this rotation sets in until for intensities higher than 7.5 mW the light pressure expels the particles from the laser focus.

Figure 4. Relative positions of laser focus and particle center. **a** For 2.80 mW laser intensity the capped colloidal particle stays close to the focus of the optical tweezers. **b** Since the successive positions follow a Boltzmann distribution, a radially averaged effective potential can be derived. **c** Seen in the reference frame of the colloidal particle, the laser spot stays close to the center. **d** At 5.44 mW particles are expelled from the center and rotate at a finite distance about the beam axis. **e** The potential shows a global minimum corresponding to rotation, while the small dip identifies normal tweezing. Typical error bars were estimated. **f** The rotation is counterclockwise for a position of the focus on the left side and clockwise for positions right to the center. The limited resolution of the optical microscope causes the grid-like distribution of data points in panels **a**, **d**, and the circular artifacts in **c**.

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